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TIME-OF-FLIGHT MEASUREMENT OF N_2^+ MEINEL LIFETIMES

SRI Project PYU-1364

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13 ABSTRACT

Radiative lifetimes of $N_2^+(A^2\Pi_u)$ have been determined for vibrational levels $v' = 1$-8. They decrease monotonically from 13.9 ± 1.0 μsec for $v' = 1$ to 7.3 ± 0.5 μsec for $v' = 8$. A time-of-flight method was used to overcome collisional and diffusion effects that have affected most other measurements.

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INTRODUCTION

This report covers the research carried out under ONR Contract N00014-71-C-0366, which was funded under ARPA Order No. 1859, Program Code 1F10. The work has been successfully completed and the results have been prepared for publication. Because this article contains a description of the work and the results, we are attaching it as the main technical part of this final report. In the interest of simplicity, the body of the report is therefore limited to an explanation of the motivation for the work, and some comments concerning the usefulness of the results.

THE RESEARCH EFFORT

This research effort was primarily experimental in nature, and was aimed at a reliable determination of the natural, or radiative, lifetimes of several vibrational levels of the $A^2\Pi_u$ state of N_2^+, which decay to the $N_2^+(X^2\Sigma_g^+)$ ground state, emitting the Meinel bands. This information is needed for the proper interpretation of Meinel band observations in the upper atmosphere, as well as for excitation studies in the laboratory, some of which are currently being carried out at Utah State University under an Air Force contract.

Due to the long natural lifetimes (near 10 μsec) of the $A^2\Pi_u$ levels, their determination can be most accurately made under the collisionless conditions that can be achieved using a time-of-flight method. Such an apparatus had been constructed in our laboratory several years ago. We had in fact made the first quantitative lifetime measurements on the Meinel system in a brief trial experiment in 1968[1]. More refined measurements were not made at that time because of lack of funds.

The present work has been carried out on essentially the same apparatus used earlier, but several important modifications were made that substantially increased its efficiency and improved the reliability of the data. The results are gratifyingly consistent. They have been analyzed by D. C. Cartwright to determine the dependence of the transition electronic moment on the $N\text{-}N^+$ internuclear separation. Dr. Cartwright's work will be published as a companion paper to ours, and the two will furnish a definitive description of the $N_2^+(A^2\Pi_u)$ radiative transition to $N_2^+(X^2\Sigma_g^+)$. Our results are in excellent agreement with measurements on $v' = 2\text{-}5$ recently made by Holland and Maier at Los Alamos, and reported[2] during the course of our work. They also used a time-of-flight method.

These results are in clear disagreement with the value of 6.7 μsec for $v' = 2$, which was obtained by O'Neil and Davidson[3] and used by Shemansky and Broadfoot[4] as a normalization point for their measurements. We obtain a value of 11.9 ± 0.4 μsec, and Holland and Maier find 12.3 ± 0.7 μsec. The exceedingly good internal consistency of our measurements over eight different vibrational levels, as well as the close agreement with Holland and Maier (who used a different time-of-flight configuration), give a high level of confidence to these results, and justify the effort spent in obtaining them.

Close communication was maintained with Dr. Cartwright during the course of the experiments, to permit him to feed new data into his analysis. This cooperation was beneficial to both efforts. Our original research goals were modified when Cartwright was able to show that an extension of the study to include higher v' levels was more important than improving the accuracy of the $v' = 1$ results. The reason is that the $v' = 0$ and 1 lifetimes depend on electronic moments for internuclear separations R that are common to the $v' = 2$ and higher levels, and the R dependence can be more accurately determined by utilizing data from higher v' levels. These data allow a larger range of R to be included in the analysis than would be for only the levels $v' = 1$-5. Therefore, the levels 6, 7, and 8 were also studied. The final array of data covering the levels $v' = 1$ to 8 has now been analyzed by Dr. Cartwright, who has been able to obtain a good definition of the absolute value of the electronic moment and its dependence on R.

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TIME-OF-FLIGHT DETERMINATION OF LIFETIMES* OF $N_2^+(A^2\Pi_u)$--THE MEINEL BAND SYSTEM

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ABSTRACT

Radiative lifetimes of $N_2^+(A^2\Pi_u)$ have been determined for vibrational levels $v' = 1$-8. They decrease monotonically from 13.9 ± 1.0 μsec for $v' = 1$ to 7.3 ± 0.5 μsec for $v' = 8$. A time-of-flight method was used to overcome collisional and diffusion effects that have affected most other measurements.

INTRODUCTION

Since its discovery in a natural aurora in 1950[1], the Meinel band system $N_2^+(A^2\Pi_u \rightarrow X^2\Sigma_g^+)$ has become recognized as an important red and near infrared component of auroral and other airglow emission spectra. Although the molecular constants of the $A^2\Pi_u$ state are by now fairly well established, the radiative and collisional properties are not. Quantities such as natural lifetimes, excitation cross sections, and collisional quenching rate constants determine the flourescence efficiency and are useful in interpreting airglow emission intensities in terms of the excitation conditions. In spite of a considerable amount of effort, laboratory experiments have generally failed to give consistent values for the excitation cross sections and the natural lifetimes of the various bands. The difficulties in laboratory measurements, which usually employ electron beams to excite N_2 gas, can ultimately be attributed to either the long natural lifetime and attendant collisional quenching and space charge or diffusion effects, or in some cases, to interference from overlapping bands of the N_2 first positive system.

In 1963 a value in the vicinity of 3 μsec was first tentatively placed on the natural lifetime by Sheridan et al.[2], from an interpretation of the spatial diffuseness of Meinel band emissions compared to those of the N_2^+ first negative system excited by a proton beam. Subsequently, O'Neil and Davidson[3] obtained values near 6 μsec using a pulsed electron beam method. However, Hollstein et al.[4] in this laboratory obtained values near 12 μsec using a time-of-flight method. Barring some unusual cascade contribution in the latter measurements, which were carried out under collision-free conditions, the discrepancy would be attributed to collisional quenching or diffusion losses in the pulsed beam experiment.

When natural radiative lifetimes are in the 10 μsec range, either collisional quenching or diffusion losses (or both) are usually present at the pressures and in the observation volumes in which excited gases are studied in the laboratory. These problems are discussed in a recent paper by Shemansky and Broadfoot[5], who note that the variations (by a factor of two or more) in the values of excitation cross sections reported by different workers can be attributed not only to collisional effects but also to the effects of various electric fields on the $N_2^+(A^2\Pi_u)$ densities in the observation region. They also noted that Meinel measurements are further complicated, when using electron beams to excite the gas molecules, by the fact that some bands are overlapped by bands of the N_2 first positive system, which is simultaneously excited.

Measurements using time-of-flight techniques do not suffer from any of the above shortcomings. There are some problems related to intensities, beam focusing, and so forth, but there are no extraneous losses resulting from collisional quenching or diffusion, and there is no interference from first positive bands in a pure N_2^+ beam. Because our original time-of-flight measurements[4] were made in a short exploratory study and were considered to be preliminary in nature, and because of the general importance of the N_2^+ Meinel system in atmospheric emissions and the lack of agreement that has existed among the different laboratory studies, we have made new measurements on the system, which we report here. A time-of-flight method similar to our original one was used. However, apparatus improvements and more extensive data have greatly increased the reliability and precision of the results. Furthermore, a total of eight v′ levels were studied to permit a definitive description of the absolute value of the transition moment M(R) as a function of internuclear separation R. An analysis of the data by D. C. Cartwright in the companion paper[6] demonstrates the internal

consistency of the results. During the course of our work, Holland and Maier[7] completed measurements on four vibrational levels of $N_2^+(A^2\Pi_u)$. They also used a time-of-flight method, but in a different configuration than ours. Their results agree exceedingly well with those reported here.

Because the theoretical relationships between the natural lifetimes, transition probabilities, and the electronic moments have been extensively discussed elsewhere, and are further discussed and referenced in the companion paper by Cartwright, we avoid the redundancy of repeating them here. We merely discuss our experimental work and the results.

EXPERIMENTAL METHOD AND PROCEDURE

Apparatus

The lifetime measurements were made by observing the optical emissions from particular vibrational levels of the $A^2\Pi_u$ state of N_2^+ as a function of the ion time-of-flight between two observation points. The apparatus used is shown in Figure 1. N_2^+ ions are produced in a hot-filament dc discharge by electrons with a maximum energy of 100 eV. A substantial fraction of the N_2^+ produced by this source is in the $A^2\Pi_u$ state. The ions are accelerated to the desired energy, mass selected, and directed into the observation chamber. The intermediate chamber in Figure 1 contains a charge transfer cell, which is used for measurements on neutral species.[4,8,9] To increase the N_2^+ intensity in the observation region, the beam path length from the source was shortened by removing this chamber for most of the measurements reported here; the observation chamber was attached directly to the mass analyzer chamber.

In the observation chamber, two identical front surface mirrors, oriented at 45° and 135° to the beam direction (see Figure 1), sample light from two 2-cm segments of the beam at positions separated by 15.3 cm. Centered between these mirrors is a third, larger mirror that can be rotated to direct the light from either sampling mirror vertically through a quartz lens and a quartz window in the vacuum wall, through an optical interference filter, and onto the cathode of a Freon-cooled photomultiplier tube (RCA31000E). The relative light intensity from each beam segment is determined by counting output pulses from the photomultiplier.

The total ion current is monitored at the end of the observation chamber by a double collector arrangement. The first collector has an oval aperture 0.7-cm wide and 1.5-cm high that is centered on the beam axis. Maximizing the current to the second collector so that most of the beam passes through the aperture in the first collector assures that the beam is properly directed and collimated. For most of the measurements reported here it was possible to focus and direct the beam so that 70 to 90% of the total beam reached the second collector. The observation apertures are 2-cm wide, assuring that essentially all of the beam was observed at both apertures. A honeycomb light baffle is located just in front of the beam collector to prevent fluorescence at the collector surfaces from producing an optical signal.

Method

The lifetime τ for a particular vibrational level is determined from the relation[8]

$$\tau = -\frac{b}{v}\left[\ln(I_1/I_2) + \ln \alpha\right]^{-1} \qquad (1)$$

where v is the beam velocity, b is the distance between observation points, I_1 and I_2 are the observed photon intensities from the two beam segments, and α is the ratio of the photon collection efficiencies for the two light paths. If data are obtained over a range of velocities, then a plot of $\ln(I_2/I_1)$ vs. b/v yields a straight line of slop $(-1/\tau)$ and intercept $(-\ln \alpha)$.

For a given transition, data for I_1 and I_2 were obtained over the velocity range from 3×10^6 cm/sec to 12×10^6 cm/sec. At each velocity, 10 to 100 separate observations of I_1 and I_2 were made. To obtain I_1

and I_2, a background signal, obtained when the ion beam was deflected just before entering the observation chamber, must be subtracted from the total signal. This background was almost entirely due to dark counts from the photomultiplier, but the subtraction also accounted for any signal resulting from light sources other than the ion beam. The background count rate was typically near 25/sec. Total counting rates ranged from 50/sec to 10^4/sec. The counts were taken over equal time intervals ranging from 10 to 300 sec, depending on the count rate, which varied with beam energy and the particular band being observed.

From the values of I_1 and I_2, the ratio I_2/I_1 and its standard deviation were calculated. This procedure was repeated for each of 10 to 16 velocities, and then a computer was used to determine a least-squares fit to the equation

$$\ln(I_2/I_1) = -\frac{1}{\tau}\frac{b}{v} - \ln \alpha \quad . \tag{2}$$

Illustrative data are shown in Figure 2, which gives a semilog plot of I_2/I_1, versus b/v, for (3,1) band data. The straight line represents the computer fit. In the fitting procedure each point was weighted by the inverse square of its standard deviation. The statistical errors in the coefficients τ^{-1} and $\ln \alpha$ were calculated using standard statistical procedures.[10] In this method, α is treated as a parameter that is assumed to remain constant during a set of measurements on a given band. It was found that α could be altered by a physical change along either the separate or the common light paths. If any such change was made, the new data were handled separately and a new value of α was sought. It was interesting to note that α often changed by a few percent when only the interference filter was changed for the study of a different band. Apparently the nonuniformity of the photocathode was wavelength dependent, and the

beam images from the two sampled sections (resulting primarily from optical asymmetries) were sufficiently different to cause the net change in α. Nevertheless, during a series of measurements on a given band over a wide range of energies (150 to 2000 eV) α remained constant, as can be seen from the linearity of the data in Figure 2. The double-collector system mentioned above was designed to facilitate the focusing to collimate the beam well at all energies so that the beams always had similar physical shapes, and thus to stabilize α for each set of measurements at a particular wavelength. During the course of all these measurements, α remained between 0.90 and 0.96, except for the (1,0) measurements where its value was 1.10. The (1,0) bandhead wavelength of 9182 $\mathring{A}$ is near the long wavelength limit of the photocathode sensitivity. In this wavelength region the average quantum efficiency is strongly dependent on wavelength, and it is also, apparently, subject to local variations across the surface of the photocathode. These factors caused α, for the (1,0) measurements, to lie outside the normal range of values. This threshold characteristic could also render α very sensitive to the intensity distributions of the beam images on the photocathode, and may have added to the fluctuations in the data obtained for the (1,0) band.

Our two-mirror technique effectively normalized the signal at the second aperture to that at the first, and observed the decay of the A state levels as a function of transit time from the first to the second aperture. An alternative procedure is to observe the photon intensity I at a fixed single position as a function of beam velocity. If it is assumed that the fractional population of a given vibrational level is directly proportional to the total ion beam current i_T, then the lifetime can be determined from the slope of a plot of ln (vI/i_T) vs. b/v. That is the method employed by Holland and Maier.[7] Tests with the ion source used in this research revealed that under a wide variety of source conditions the fraction of ions extracted in the $A^2\Pi_u$ levels was a function of the discharge voltage.

Further, the residence time of an ion inside the source before extraction is unknown, and may contribute a significant error. We observed that the population of the $A^2\Pi_u$ levels relative to the total ion current did not remain constant as the extraction voltage was varied. None of these uncertainties arise in the two-mirror technique, and as a result all measurements reported here were obtained using this method.

Uncertainties

With the technique used, possible sources of systematic error are minimized. Only the distance between observation points and the beam velocity can contribute to systematic errors. The distance is 15.3 $\pm$ 0.05 cm. Retarding potential measurements of the beam energy and energy spread showed that the uncertainty in the energy of the ions was always less than 1% of the mean beam energy. These two factors can contribute at most a $\pm$ 0.1 μsec uncertainty to the lifetime measurements.

The statistical errors depend on the intensity of the band under consideration, the background noise level, fluctuations in the ion current, and possible changes in the counting electronics. The magnitudes of these uncertainties were determined from the data distributions using standard techniques[10] and are consistent with observed variations in the results.

Results

The results are presented in Table 1, which also lists the particular band studied for each v' level, and the wavelength of the bandhead. The lifetimes range from almost 14 μsec for $v' = 1$, to 7.3 μsec for $v' = 8$, and are consistent with a smoothly varying dependence of the $A^2\Pi_u - X^2\Pi_g$

electronic moment on internuclear separation.[6] For $v' = 5$, both the (5,1) and (5,2) bands were measured; the two results were in good agreement, and have been combined in Table 1. Sufficient data were taken on most bands to reduce combined statistical and estimated systematic errors to 0.5 μsec or less (as is indicated in the table). The $v' = 1$ level was the exception. As a result of a low production efficiency in the source (due mainly to a small Franck-Condon factor from ground state $N_2(X^2\Sigma_g^+, v=0)$) and low photocathode quantum yield near 9182 Å, the counting rates I_1 and I_2 were very low and sufficient data were acquired only to reduce the combined uncertainties to 1.0 μsec. We had initially intended to reduce these uncertainties by acquiring more extensive data, but the analysis of Cartwright indicated that it would be more valuable to extend the measurements (which initially covered only $v' = 1$ through 5) to higher v' levels. The reason for this is that the range of electronic moments appropriate to the $v' = 0$ and 1 transitions are in the same range as those for the levels $v' = 2$ and higher. Thus, Cartwright's analysis could provide reliable values for $v' = 1$ and 0 if the other levels were well measured, and more information on the R-dependence of the electronic moment $M(R)$ could be obtained by extending the measurements to higher v' levels than by measuring the lowest ones. We therefore measured lifetimes for $v' = 6$ through 8.

Discussion

A comparison of our results with those of Holland and Maier,[7] which are also presented in Table 1, demonstrates the exceedingly good agreement obtained in the two time-of-flight experiments. It also appears that the uncertainty of 0.7 sec estimated by Holland and Maier is probably a little larger than standard deviations from the mean. With these measurements,

we feel that the natural lifetimes of the $v' = 1$ through 8 levels of $N_2^+(A^2\Pi_u)$ are reliably defined. The analysis of Cartwright[6] further establishes the lifetimes of the $v' = 0$ and 1 levels as well as the dependence of the $N_2^+(A^2\Pi_u - X^2\Sigma_g^+)$ transition moment on the internuclear separation.

Figure 3 permits a graphical comparison of most of the lifetimes that have been measured either in the laboratory or obtained by semi-empirical methods. O'Neil and Davidson's pulsed electron beam study[3] was mentioned earlier. Shemansky and Broadfoot[5] measured electron-impact excitation cross sections and relative band intensities under electron beam excitation, and used the latter, with RKR Franck-Condon factors, to calculate the variation of electronic moment with R. However, some low pressure anomalies connected with quenching, diffusion, and space charge effects prevented them from obtaining the radiative lifetimes, so they normalized their relative values to the results of O'Neil and Davidson.[3] Popkie and Hennecker[11] used the relative band intensities obtained (under electron beam excitation) by Stanton and St. John [12] to determine the R dependence of the transition moment, and used experimental lifetime data of O'Neil and Davidson[3] and also the preliminary time-of-flight measurements[4] to establish the absolute magnitude. Only those measurements based on the latter data are shown in Figure 3. Their lifetimes fall off relatively more rapidly with increasing v' than the present results. The value of 9.2 ± 2.0 μsec for $v' = 2$ was recently obtained by Gray, Morack, and Roberts[13] using pulsed electron beam excitation. The results of Holland and Maier[7] for $v' = 1$-5 are in clear agreement with this work. Their more recent measurement[14] of 6.2 ± 0.4 μsec for $v' = 10$ is also apparently consistent.

It is clear that all pulsed electron beam measurements have yielded lifetimes that are shorter than the natural $N_2^+(A^2\Pi_u)$ lifetimes, because of collisional quenching or diffusion losses of the type described earlier.

At the low pressure limit of such experiments, the thermal ions move about 0.5 cm in one radiative lifetime without the influence of space charge fields, and in most apparatuses this is a sizable fraction of the distance from the beam axis to the edge of the observable volume. Thus, normal diffusion will usually shorten the lifetimes at low pressures, and the effects become more complex when space charge forces are included. Consequently, many of the electron beam experiments should probably be reinterpreted. Clearly, the results should be reexamined that led Shemansky and Broadfoot to conclude[5] that lifetimes as long as these were inconsistent with their excitation cross sections. The determinations of electron-impact excitation cross sections by Holland and Maier[7] are doubtless consistent with these results in view of their agreement on the lifetimes. However, their technique is not ideal for obtaining absolute excitation cross sections. Thus, further excitation and quenching measurements using electron beam excitation should be carried out in experiments that are designed with the knowledge of these radiative lifetimes. One experiment is currently underway at Utah State University.[14]

These lifetimes, when combined with accurate quenching and excitation cross sections, should enable considerably more reliable interpretations of airglow measurements to be made than are now possible. They also indicate that $N_2^+(A^2\Pi_u)$ may often be present in ion beams produced in conventional ion sources. The long natural lifetimes plus the high initial ion fraction (Holland and Maier[7] estimate that 40% of the ions produced by 100-eV electrons are in $A^2\Pi_u$) are responsible. N_2^+ beam experiments in which the flight times to the interaction (or observation) area are less than 20 μsec should be analyzed with this possibility in mind.

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Table 1

$N_2^+(A^2\Pi_u)$ MEINEL LIFETIMES

Vibrational Level of N_2^+ A State	Band	Bandhead Wavelength	Lifetime τ (μsec)	
			This Research	Holland and Maier[7]
v'	(v',v")	Å		
1	(1,0)	9183	13.9 ± 1.0	
2	(2,0)	7854	11.9 ± 0.4	12.28 ± 0.7
3	(3,1)	8083	10.7 ± 0.4	10.70 ± 0.7
4	(4,0)	6124		10.08 ± 0.7
	(4,1)	7065	9.7 ± 0.4	
5	(5,1)	7264	9.1 ± 0.4*	
	(5,2)	6286		9.14 ± 0.7
6	(6,3)	7473	8.4 ± 0.5	
7	(7,3)	6634	7.8 ± 0.5	
8	(8,3)	5974	7.3 ± 0.5	

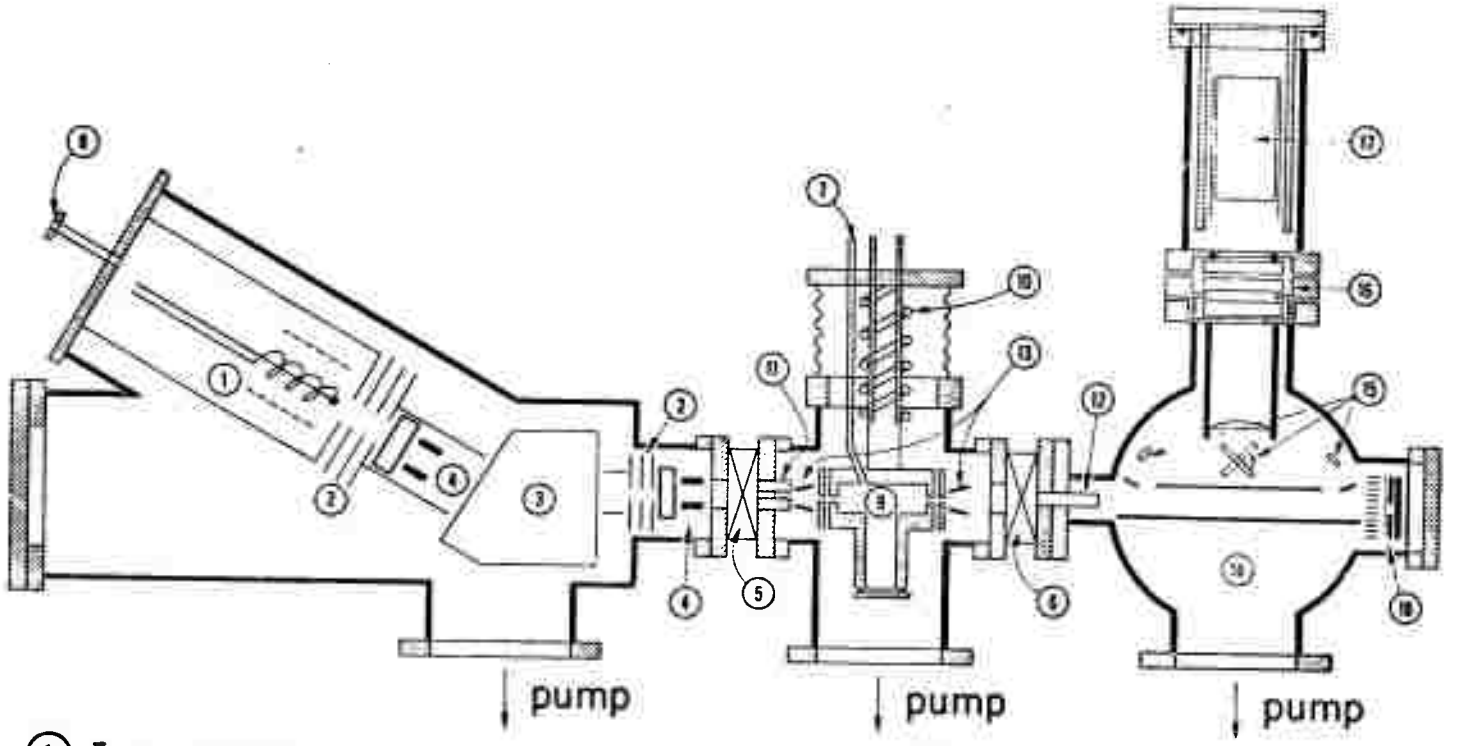
*Combined results from measurements on (5,1) and (5,2)

FIGURE CAPTIONS

1. Diagram of the apparatus. The central section containing the charge transfer oven is normally used for the production of excited neutral beams and was removed for most of these measurements.

2. Plot of the data taken for the (3,1) band. The curve is a least-squares fit to the data points and yields τ = 10.7 μsec. The data show a 0.3 μsec standard deviation from the mean, which has been increased by 0.1 μsec to include possible systematic errors.

3. Comparison of lifetime determinations for the N_2^+ Meinel System.



① Ion source
② Lens system & deflecting plates
③ Magnet
④ Directional adjust
⑤, ⑥ Gate valves
⑦, ⑧ Gas inlets
⑨ Charge transfer oven
⑩ Cooling
⑪, ⑫ Apertures
⑬ Deflecting plates
⑭ Observation Chamber
⑮ Mirrors
⑯ Interference filter
⑰ Photo multiplier
⑱ Current collector

TD-5962-13R

Figure 1.

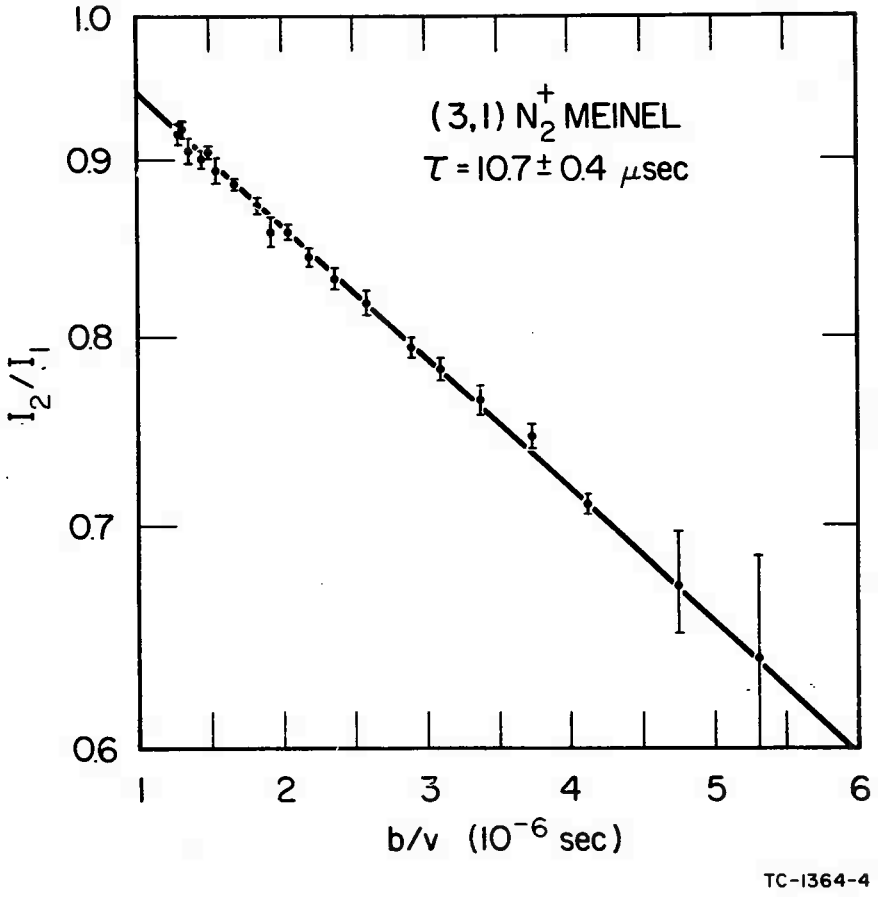

1.0
0.9
0.8
0.7
0.6
I_2/I_1
(3,1) N_2^+ MEINEL
$\tau = 10.7 \pm 0.4$ μsec
1
2
3
4
5
6
b/v (10^{-6} sec)
TC-1364-4

Figure 2.

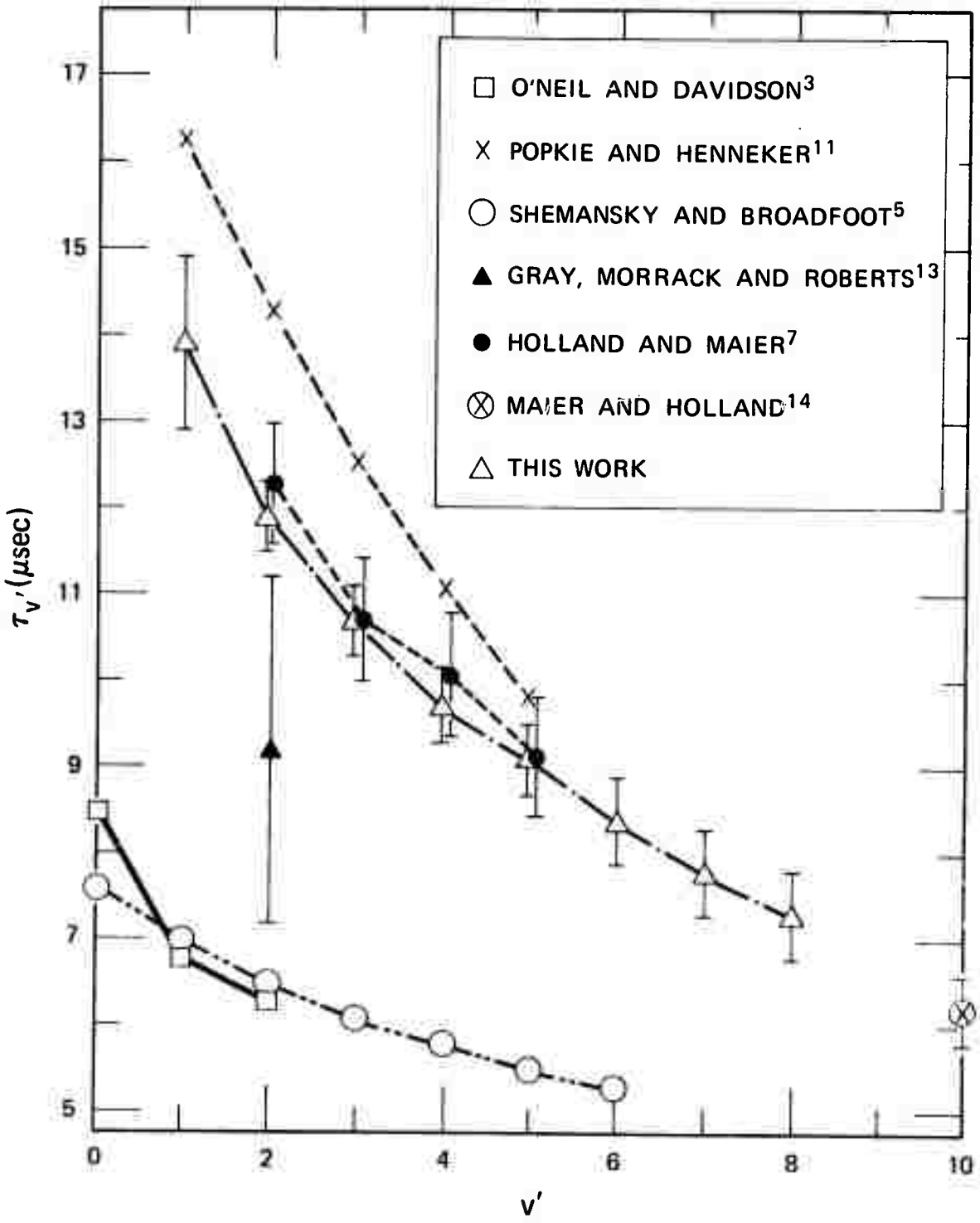

□ O'NEIL AND DAVIDSON[3]
X POPKIE AND HENNEKER[11]
○ SHEMANSKY AND BROADFOOT[5]
▲ GRAY, MORRACK AND ROBERTS[13]
● HOLLAND AND MAIER[7]
⊗ MAIER AND HOLLAND[14]
△ THIS WORK
$\tau_{v'}$(μsec)
v'
17
15
13
11
9
7
5
0
2
4
6
8
10

Figure 3.